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Improvements of FRAM version 6.1

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ABSTRACT

Medium-resolution gamma-ray spectrometer (MRGS) detectors such as the LaBr₃ and CZT detectors present a more practical and less expensive alternative to high-resolution gamma-ray spectrometers because they operate at room temperature, are more compact and reliable, and are easy to use and maintain. Making them capable of isotopic analysis of uranium and plutonium samples is a desirable feature, which is useful in various safeguards applications. That feature is provided by the upgraded Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) code, which can analyze the isotopic compositions of uranium and plutonium from spectra obtained by a LaBr₃ or CZT detector system. This upgraded code is FRAM version 6.1, to be released soon. It is a major upgrade from the current version 5.2, which was released in 2013. In addition to the capability to analyze the LaBr₃ and CZT spectra, FRAM v.6.1 will implement various new features to improve usability, sustainability, compatibility, and maintainability.

A. INTRODUCTION

The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) software was developed and continues to be refined by Los Alamos National Laboratory. It was first developed in the mid-1980s, running on a MicroVAX computer with the VMS operating system [1]. The first version of the FRAM code running on PC/Windows 3.1, called PC/FRAM, was released in 1994 [2, 3].

FRAM version 2 was the first commercially available version, released in 1997, and it ran on Windows 3.1 and 95. It was licensed to Ortec [4].

FRAM version 3 was released in 1999 as a 32-bit code, running on Windows 95, 98, and NT. It was licensed to Ortec and Canberra [5].

FRAM version 4 was released in late 2001 and early 2002 and was last updated with some bug fixes in October 2002 [6]. It had the capability to analyze the 100-keV X-ray region of both plutonium and uranium spectra. It ran on Windows 95, 98, NT, 2000, and XP. It was licensed to Ortec and Canberra.

FRAM version 5.1 was released in 2011 with a completely different user interface and the capability to run in the command line mode. It ran on all 32-bit and 64-bit Windows platforms from Windows 95 to Windows 10 [7]. It was licensed to Ortec and Canberra. FRAM v.5.2 was released in 2013 with some minor upgrades from v.5.1 and was also licensed to Ortec and Canberra.

We are working on an upgraded FRAM and planning to release it as FRAM version 6.1 in late 2019. The major upgrade of FRAM v.6.1 is its capability to analyze the LaBr₃ and CZT spectra. Additionally, FRAM v.6.1 implements various new features to improve usability, sustainability, compatibility, and maintainability. It has not been extensively tested, but it should be able to run on Windows 7, 8, and 10 and on most Windows XP operating systems.

B. MAJOR IMPROVEMENT

The major improvement of FRAM v.6.1 is its capability to analyze spectra obtained by medium resolution detectors such as LaBr₃ and CTZ.

1. LaBr₃ analysis

Below are the basic steps FRAM v.5.2 uses to determine the isotopic composition:

- i. Determine energy calibration.
- ii. Determine full width at half-maximum (FWHM) formula.
- iii. Determine peak shape formula.
- iv. Fit the regions to obtain peak areas.
- v. Determine relative efficiency curve.
- vi. Calculate the relative activities of the isotopes.
- vii. Adjust the background of the analysis regions.
- viii. Repeat steps iv to vii several times (default: five iterations).
- ix. If the ²⁴²Pu or ²³⁶U isotope is not measured or declared by the operator, it can be estimated by correlation.
- x. Calculate the final isotopic fractions of the isotopes.

In FRAM v.6.1, some of the steps above have been expanded and/or modified to respond to what is needed for LaBr₃ spectrum analysis. We employ three new steps in FRAM v.6.1 (described here as steps a–c) to analyze the LaBr₃ spectra.

a. Repeated calibrations

The resolution of the HPGe detector is good, and as a result, there are many intense, standalone peaks that can be used to determine the energy calibration, the FWHM calibration, and the peak shape calibration (steps i to iii above). These calibrations are done only once, and the values obtained from these calibrations are then used as constants in the least squares fits of the peaks in step iv. For the LaBr₃, due to its broad resolution, there are no or few good peaks to use for calibrations. Therefore, we have to use overlapping peaks for calibration. The calibrations will not be good at first, but they will become more precise as the calibration process is repeated. Therefore, in step viii above, instead of “Repeat steps iv to vii several times,” the LaBr₃ analysis calls for “Repeat steps i to vii several times.”

b. Nonlinear least-squares fit

FRAM uses linear least squares to fit the peaks of the HPGe spectra in step iv above. For a LaBr_3 spectrum, due to its very broad peaks and the overlapping of many peaks, the linear least squares fit does not work well. We therefore use a nonlinear least squares fit technique, combining the Powell's minimization method with the linear least squares fit.

Figure 1 shows an example of the fit of the 186-keV region of a uranium spectrum. Note that due to the sparsely populated peak distribution and overlapping peaks, the uranium spectrum of a LaBr_3 detector can be fitted using a linear fit like that of an HPGe detector. The fit shown in Figure 1 was achieved using the nonlinear least-squares fit.

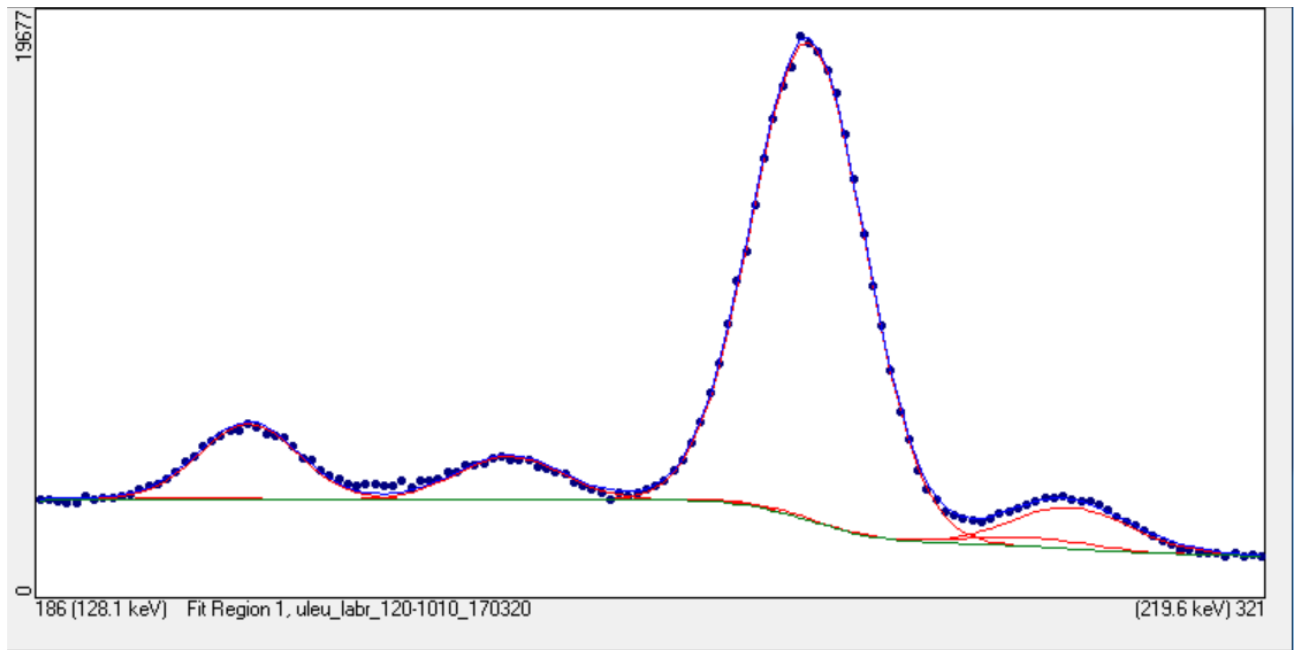


Figure 1. The fit of the 186-keV region of a uranium spectrum of a LaBr_3 detector.

Figure 2 shows an example of the fit of the region around 400 keV of a plutonium spectrum. This fitted region is about 200 keV wide, covering more than 50 measurable peaks.

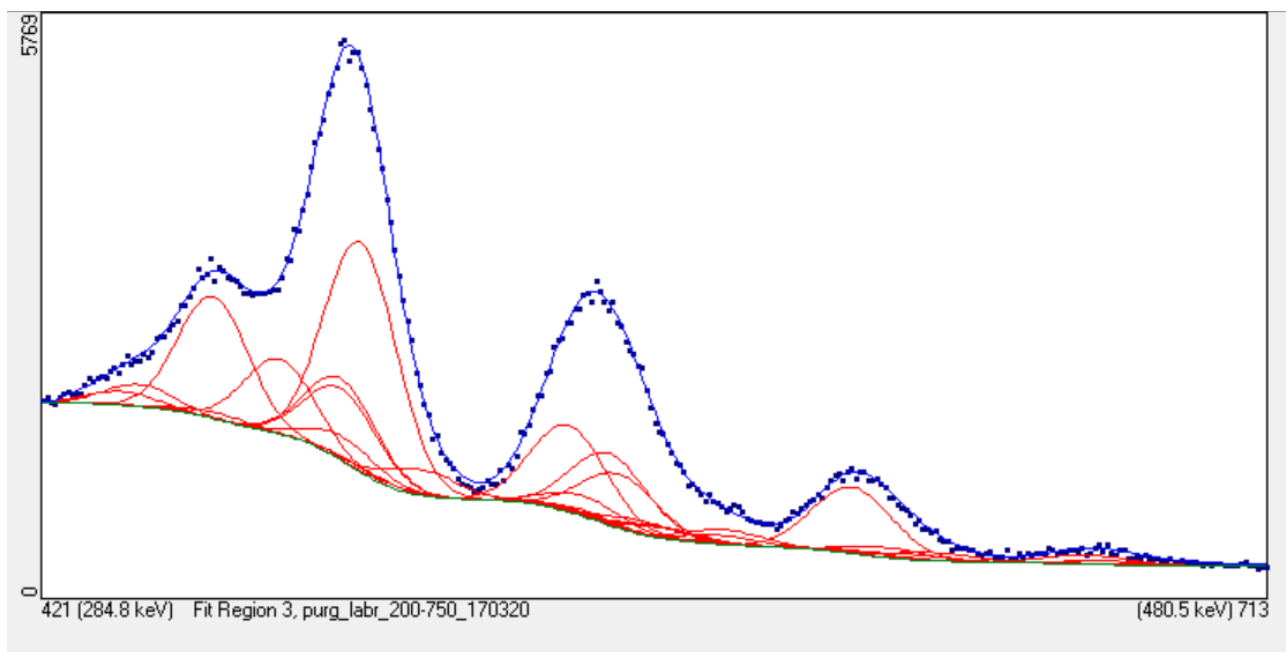


Figure 2. The fit of the 285- to 481-keV region of a plutonium spectrum of a LaBr₃ detector.

c. ²³⁸Pu and ²³⁴U correlation

The LaBr₃ spectra of weak plutonium sources have relatively large peaks from radioactive lanthanum in the detector itself at about 800 keV. Figure 3 shows the plutonium spectrum region near 766 keV.

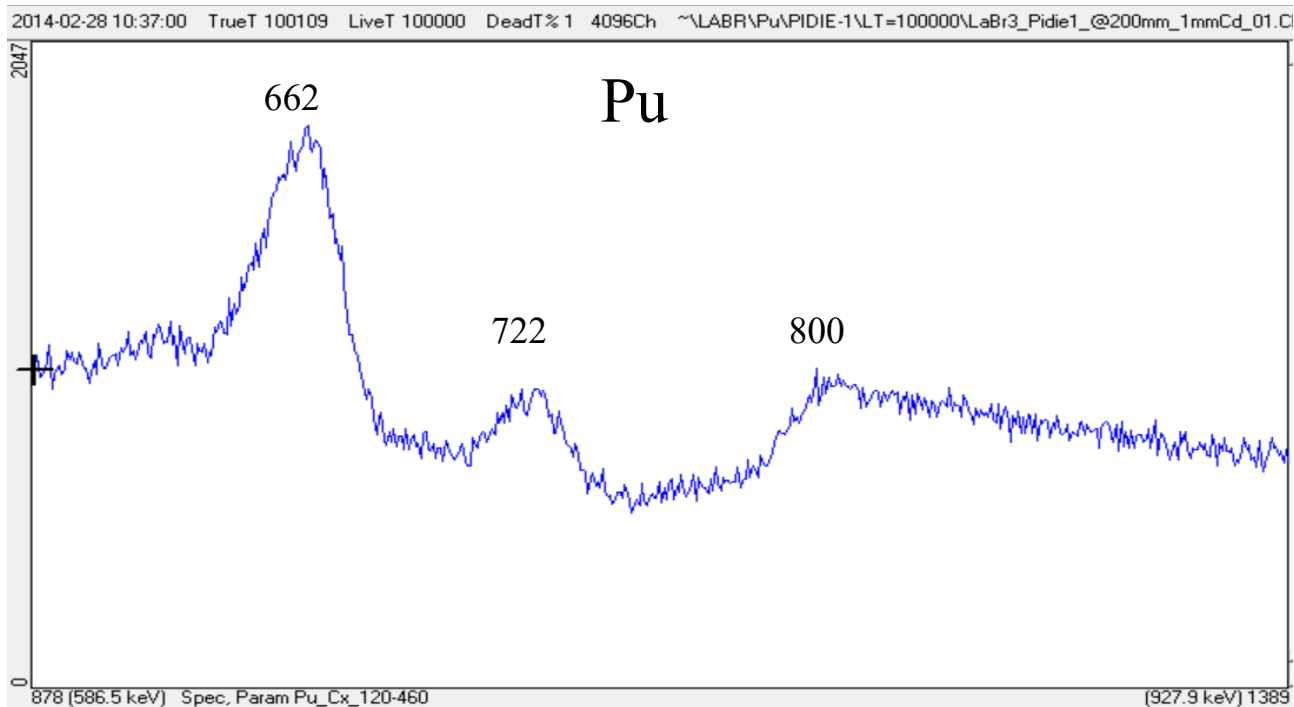


Figure 3. Plutonium spectrum region near 766 keV.

These peaks at 800 keV would interfere with the analysis in the 700- to 800-keV region and invalidate the peak area of the 766-keV peak, the only peak used to determine the activity of ^{238}Pu . Therefore, we included the option of determining ^{238}Pu activity using the correlation. The command “*Pu238_corr TRUE*” will tell the code to ignore the measured ^{238}Pu activity and will determine ^{238}Pu activity using correlation. The correlation is $\frac{^{238}\text{Pu}}{^{239}\text{Pu}} = c \left(\frac{^{240}\text{Pu}}{^{239}\text{Pu}} \right)^d$, where c and d are some constants.

Similarly, we also included the option of determining the ^{234}U activity using the correlation. It was in response to the fact that the only peak of ^{234}U at 121 keV was weak and not measurable by the LaBr_3 detector. The correlation is $\frac{^{234}\text{U}}{^{235}\text{U}} = a \frac{^{235}\text{U}}{^{238}\text{U}} + b$, where a and b are some constants. The command *P234_corr* controls how the fraction of ^{234}U is determined.

2. CZT analysis

The resolution of the CZT detector is roughly proportional to the thickness of the detector crystal. A small CZT detector will have better resolution, but due to its small size, it may not be useful because it may lack sufficient efficiency for high-energy gamma-ray peaks. The analysis employed with the LaBr_3 detector, as described in subsection 1 of this section, can also be used with a large CZT detector, for which the resolution is comparable to that of the LaBr_3 detector and the efficiency is sufficient for up to 1 MeV.

Figure 4 shows an example of the fit of the 186-keV region of a uranium spectrum obtained by the CZT-500 detector, which has a volume of 500 mm³.

Note that the peaks have very large low-energy tails and that the fit appears to address the large tails reasonably well. This capability is present in v.6.1 but not v.5.2. The description of the capability to fit a peak with very large low-energy tail will be shown in a later section of this report.

Figure 5 shows an example of the fit of the 304- to 473-keV region of the plutonium spectrum obtained by the CZT-500 detector. This region has about 50 measurable peaks.

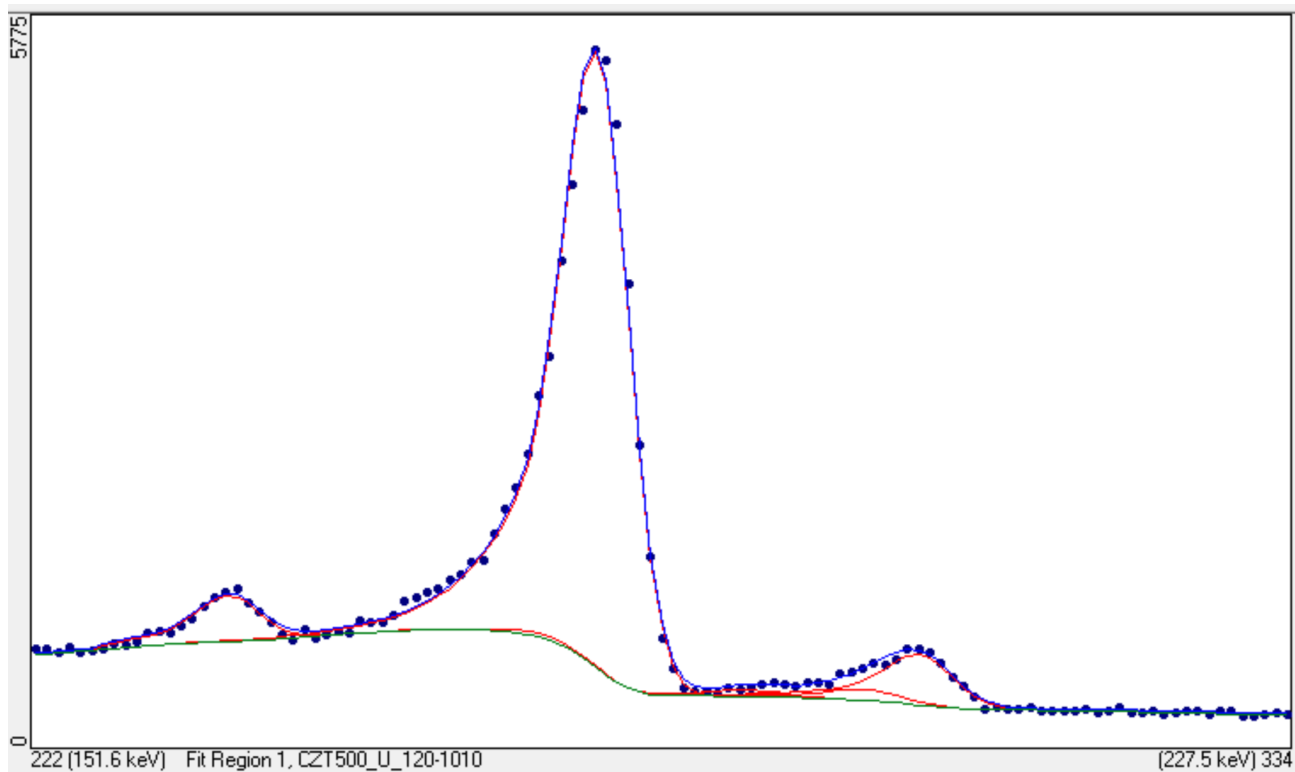


Figure 4. The fit of the 186-keV region of a uranium spectrum obtained by a CZT-500 detector.

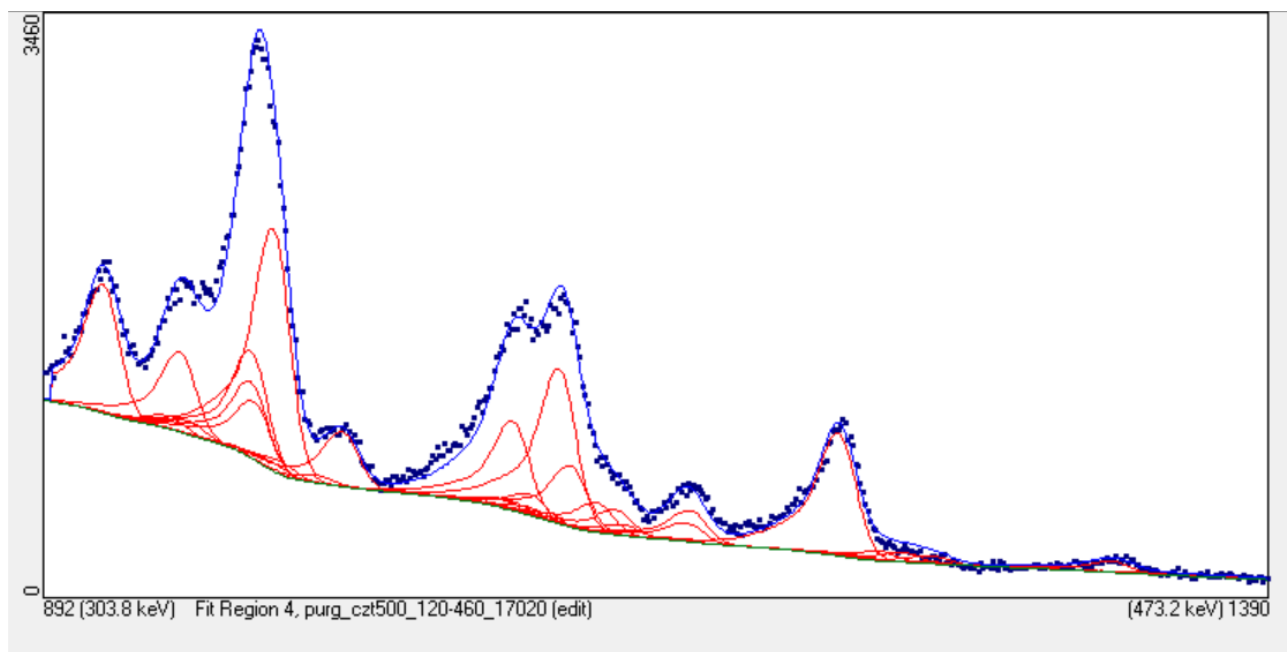


Figure 5. The fit of the 304- to 473-keV region of a plutonium spectrum obtained by the CZT-500 detector.

C. OTHER IMPROVEMENTS

Besides the capability to analyze medium-resolution spectra obtained by LaBr₃ and CZT detectors, FRAM v.6.1 also adds many new features and improvements not found on FRAM v.5.2.

1. Reading the N42 file format

FRAM v.6.1 can read the files saved with the IEEE N42 file format. This format can store multiple spectra in a file. When the N42 file format is selected, a text box appears, allowing the selection of the spectrum number in the file. Figure 6 shows an example of the N42 file format selection.

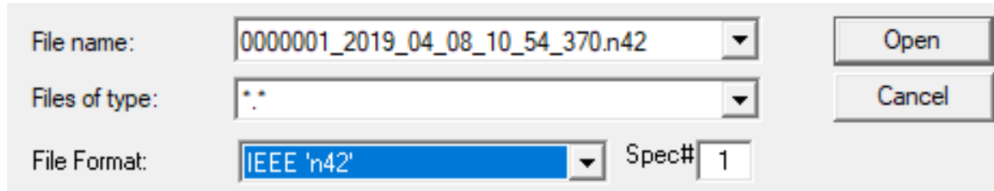
A screenshot of a file selection dialog box. It has three rows of controls. The first row is 'File name:' with a text box containing '0000001_2019_04_08_10_54_370.n42' and a dropdown arrow. The second row is 'Files of type:' with a dropdown menu showing '*. *'. The third row is 'File Format:' with a dropdown menu showing 'IEEE 'n42'' and a 'Spec#' text box with the value '1'. To the right of these controls are two buttons: 'Open' and 'Cancel'.

Figure 6. Example of IEEE N42 file format selection.

Note that FRAM can read but cannot write the N42 file format.

2. Auto file format

FRAM v.6.1 can automatically read the files that are saved with one of the following formats: Ortec 'chn', Ortec 'spc', Canberra 'mca', Canberra 'cnf', IAEA 'spe', ASCII 'txt', and IEEE 'n42'. Note that Canberra Genie 2000 would need to be installed on the computer in order to read Canberra CNF file format. Figure 7 shows an example of the Auto file format selection.

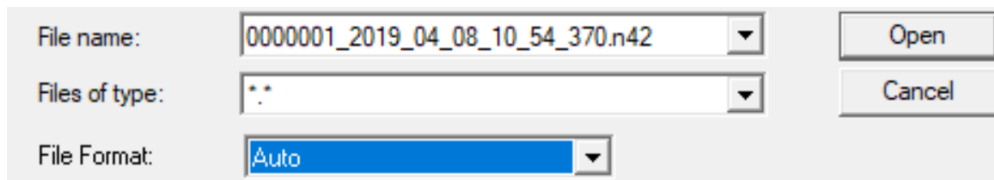
A screenshot of a file selection dialog box, similar to Figure 6. The 'File name:' text box contains '0000001_2019_04_08_10_54_370.n42'. The 'Files of type:' dropdown shows '*. *'. The 'File Format:' dropdown now shows 'Auto'. The 'Spec#' text box is empty. The 'Open' and 'Cancel' buttons are on the right.

Figure 7. Example of Auto file format selection.

When the Auto file format is selected, FRAM will try to determine the format of the file and then use the corresponding format to read the file. In some rare cases, it may incorrectly determine the file format and use the wrong format to open a spectrum. In that case, the user can specifically force FRAM to use the correct format by selecting the correct format. If the format is IEEE N42, the Auto file format will read the first spectrum in the file.

This Auto file format feature is more useful when it is used in combination with the Drag and Drop feature.

3. Drag and drop

FRAM v.6.1 allows a file to be dragged and dropped into the FRAM window and opens it using the Auto format option.

4. Sum spectra

Very often, the user measures multiple spectra of the same item in order to determine the standard deviation of the measurements and/or results from multiple spectra. The capability to sum all those spectra into a spectrum with large acquisition time is a plus. The Measure | Sum option will open the Sum dialog to allow the user to enter information about the spectra to be summed. Figure 8 shows an example of the Sum dialog box.

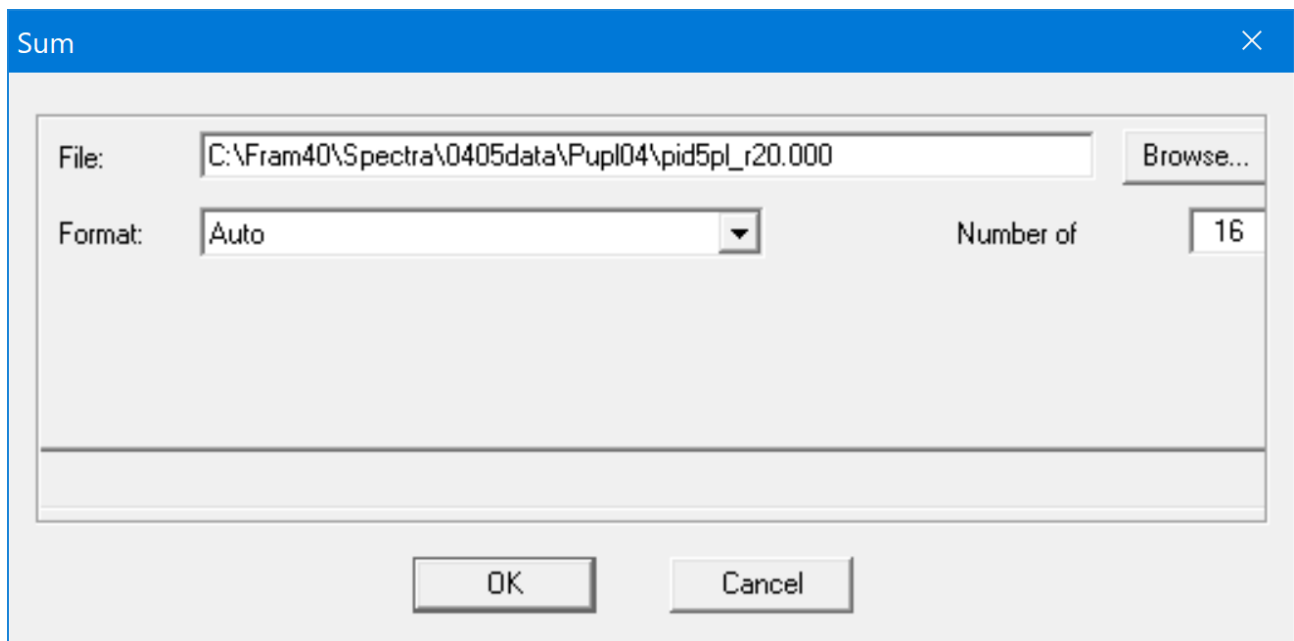


Figure 8. Example of the Sum dialog box.

5. Shift spectrum

FRAM fits a peak best when its FWHM is 8–10 channels. Many spectra are acquired with FWHM from about 20 to 100 channels wide. A weak spectrum with such wide FWHM will have many channels with zero counts. FRAM can analyze peaks when many channels contain zero counts, but the results may have large biases. It is better to compress the spectrum such that the typical FWHM is 8–10 channels. The results will be better.

The Measure | Shift option will open the Shift dialog to allow the user to enter information about the spectra to be shifted. Figure 9 shows an example of the Shift dialog box.

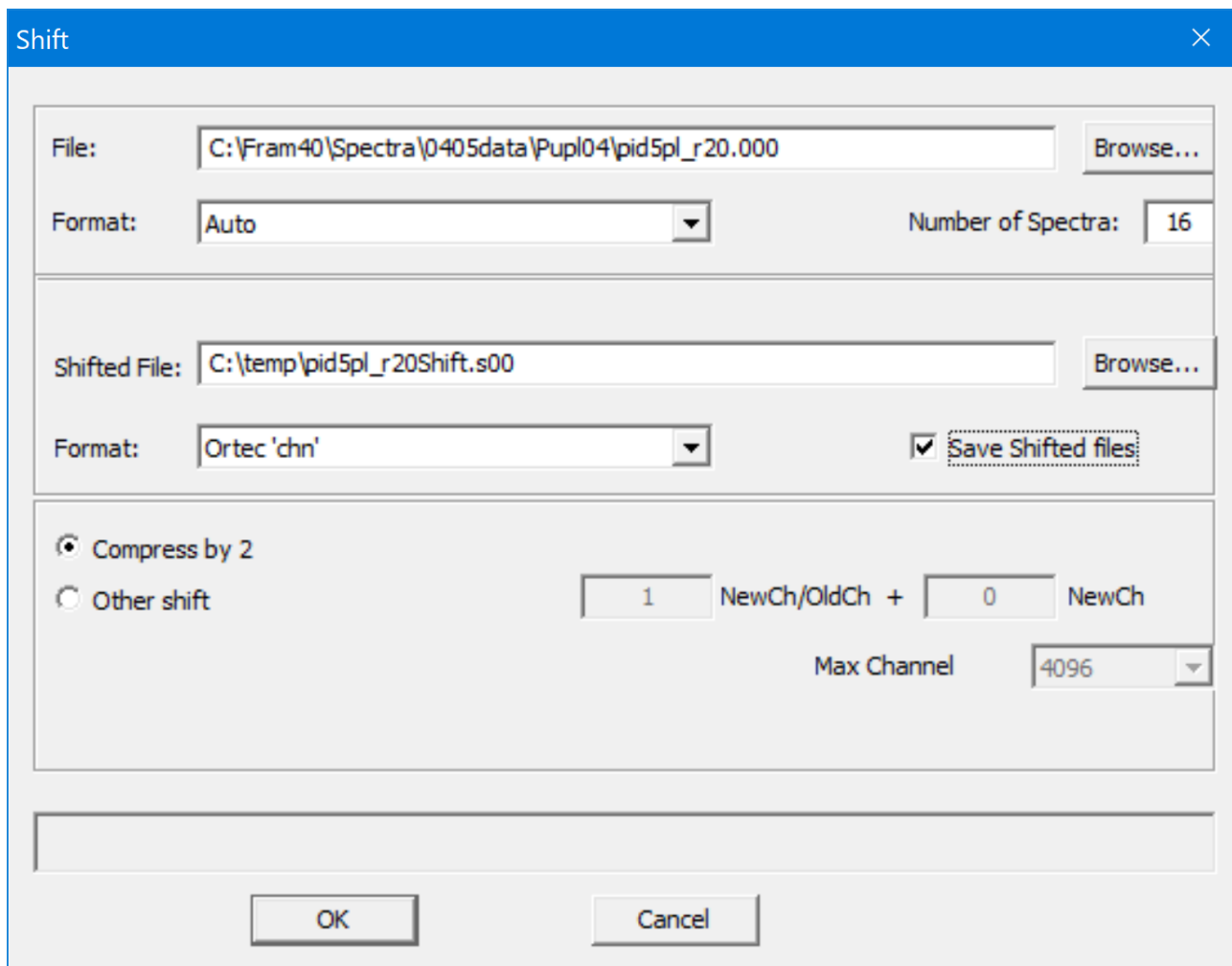


Figure 9. Example of the Shift dialog box.

The Shift option can also be used to convert multiple spectra from one format to another by checking the “Other shift” radio button and entering 1 into the NewCh/OldCh textbox and 0 into the NewCh textbox.

6. Broaden spectrum

During the course of developing the code to analyze the LaBr₃ spectra, we wrote the “Broaden spectrum” routine to assist with the analysis. The purpose was to analyze an HPGe spectrum and obtain the peak areas of all the peaks. The spectrum is then broadened so that the resolution is similar to the resolution of the LaBr₃ detector. The broadened spectrum is then analyzed with the LaBr₃ code, and the peak areas are compared with the peak areas of the HPGe spectrum.

The Measure | Broaden option will open the Broaden dialog box to allow the user to enter information about the spectra to be broadened. Figure 10 shows an example of the Broaden dialog box.

Broaden

File:

Format: Number of Spectra:

Shifted File:

Format: ☒

FWHM=sqrt(A1+A2*Ch) A1= A2=

☒ Statistically Distributed

Figure 10. Example of the Broaden dialog box.

If the “Statistically Distributed” check box is unchecked, the result will be a smoothed spectrum.

7. BG subtraction

When the background (BG) subtraction option is selected, the BG text box will appear, followed by a Browse button, allowing the user to select the BG file to be subtracted from the spectrum. This is shown in Figure 11. The file format of the BG spectrum is set to Auto and is not shown.

The parameter set for the BG spectrum is the same as that of the main spectrum. FRAM will at first read the main spectrum and calculate the energy, FWHM, and peak shape calibrations. These calibrations will be used for both the main and BG spectra. The code then reads the BG spectrum and fits the regions to determine the areas of the peaks using the calibrations obtained earlier for the main spectrum. Since the BG spectrum is taken at a different time and at a different input rate, it is likely that the energy calibration will change slightly from the main spectrum. So for the fit of a region of

the BG spectrum, the centroids of the peaks are not fixed but allowed to move together within the bounds from the fixed positions determined by the energy calibration. The bounds are $\pm (0.125 + 0.0001 * \text{Center})$, where Center is the mid-point of the region (in channel). The areas of the BG peaks are then stored in memory and subtracted from the peak areas of the main spectrum during the analysis of the main spectrum. The peak areas of the BG spectrum are displayed in the medium and long results displays.

The image shows a software dialog box titled "Analyze". It contains several input fields and checkboxes. The "File:" field is set to "C:\FRAMOPT\sorted_data\pu_coax_180-1010_long\CBNM93.SUM". The "Format:" dropdown is set to "Auto". The "Number of Spectra:" is set to "1". The "Parameter:" dropdown is set to "GeCoax_Pu_180-1010", with "0.125 KeV/Ch + 0 KeV" displayed next to it. The "Comment:" field is empty. There are three radio buttons for "Pu242/U236 by correlation", "Pu242/U236 by operator entry", and "Pu242/U236 by measurement", with the first one selected. There are also two radio buttons for "Empirical Efficiency" and "Physical Efficiency", with the second one selected. A button labeled "Efficiency defaults" is next to them. Below these are checkboxes for "Save Results" and "Print Results", both of which are unchecked. The "Result File:" field is empty, with a "Browse..." button next to it. At the bottom of the main section, there are checkboxes for "Auto analysis", "Fresh Uranium", "Previous Calorimetric measurement", and "Previous Neutron measurement", all of which are unchecked. A checkbox labeled "Subtract BG" is checked and is highlighted with a red rectangle. Below this section, there is a "BG File:" field set to "C:\temp\Area G Background.CHN", which is also highlighted with a red rectangle, along with a "Browse..." button. At the very bottom are "OK" and "Cancel" buttons.

Figure 11. BG subtraction selection option.

The BG subtraction option is available in both the Analyze and Measure dialog windows. It works with high-resolution spectra only and is ignored if the *Med-Resolution* command is set to *TRUE*.

8. Instantaneous Simulation

FRAM v.5 has the Simulation MCA in the Acquire and Measurement windows. This option generates data from a previously acquired spectrum using the Poisson distribution. The simulation clock is run at 10 times the rate of the actual clock in order to shorten the waiting time for completion of the acquisition. In addition to the Simulation MCA, version 6.1 has the Instantaneous Simulation MCA. This option instantly generates data from a previously measured spectrum. Figure 12 shows the Acquire window with the Instantaneous simulation option.

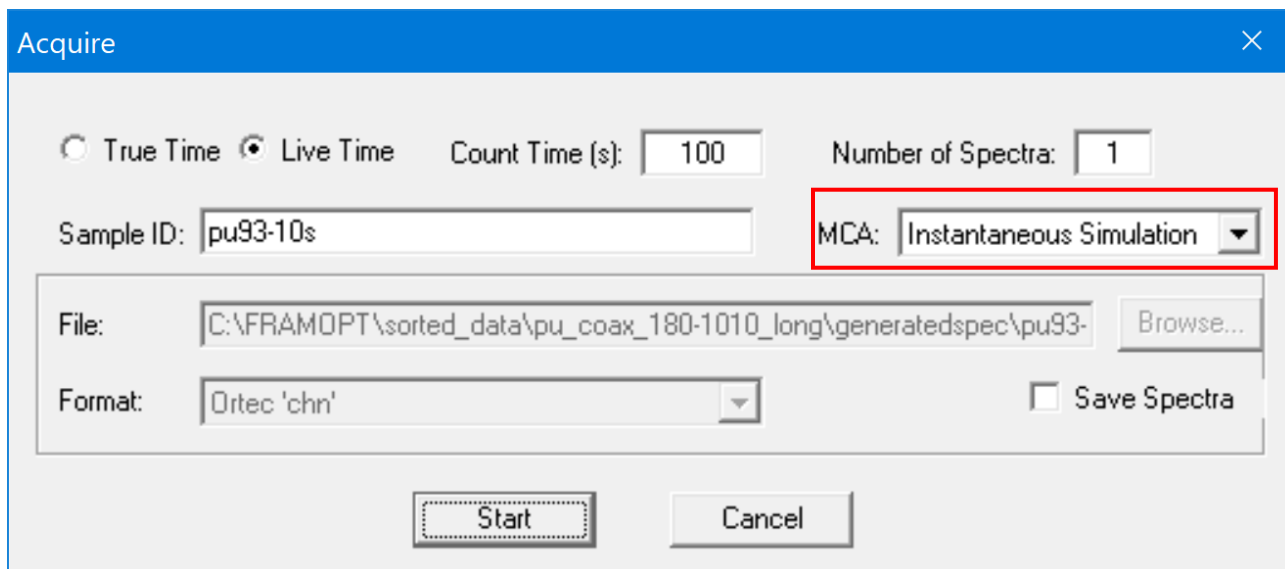


Figure 12. The Instantaneous Simulation option in the Acquire dialog window.

9. Summed peak correction for uranium analysis employing the X-ray regions

FRAM v.5 has the command *correct_u_sumpeaks* to correct for the intensities of the ^{238}U peaks that undergo coincidence summing in the analysis employing the energy region above the X-rays. This command has no effect on analysis employing the X-ray region in FRAM v.5.

U-235 decays to ^{231}Th . The decay process releases the intrinsic ^{231}Th X-rays. The uranium analysis employing the low-energy region uses these ^{231}Th X-rays. These ^{231}Th intrinsic X-rays from the decay of ^{235}U are in coincidence with many gamma rays from the decay. As a result, the ^{231}Th X-rays suffer coincidence summing and will record fewer counts in the peak areas. Version 6.1 includes the capability to correct the intensities of the ^{231}Th intrinsic X-rays. The command is the same as that of the higher-energy-region analysis, *correct_u_sumpeaks*. The intensity of the 202-keV peak of ^{235}U [4] is used to determine the magnitude of the correction.

10. U/Pu ratio from fluorescence X-rays

All the plutonium isotopes except ^{241}Pu decay to uranium. A small fraction of ^{241}Pu also decays to uranium. These decays generate intrinsic uranium X-rays. For a mixed oxide (MOX) sample, the gamma rays, X-rays, and particles will interact with the atomic electrons of uranium and plutonium

atoms and generate uranium and plutonium fluorescence X-rays. The measured uranium X-rays include both the intrinsic and fluorescence components, while all the plutonium X-rays are from fluorescence. The uranium intrinsic X-ray component is calculated using the measured plutonium isotopic compositions and then subtracted from the total uranium X-rays. The remaining uranium fluorescence X-ray component is then compared with the plutonium X-rays and the U/Pu ratio can be determined.

The analysis using the plutonium parameter set or UPu parameter set (i.e., MOX parameter set in FRAM v.5) will automatically calculate the U/Pu ratio from uranium and plutonium fluorescence X-rays. If the uncertainty of the U/Pu ratio is < 50%, then the code will display the U/Pu result. A 6% systematic uncertainty is added to the uncertainty of the ratio. Figure 13 shows an example of the short output on an analysis with the U/Pu result from fluorescence X-rays calculations.

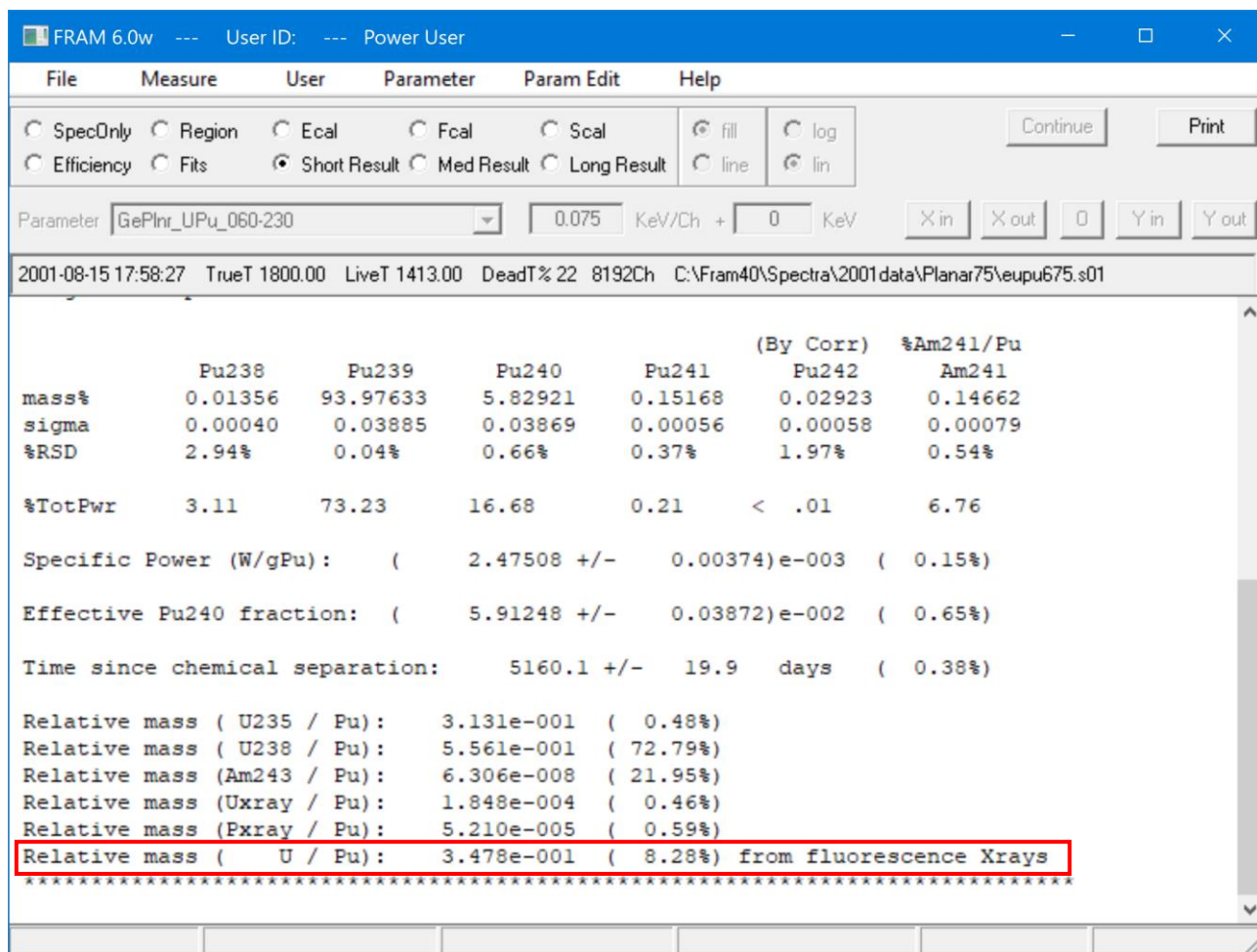


Figure 13. An example of a short output where the U/Pu ratio from fluorescence X-rays is shown.

11. CSV and log file

After an analysis, FRAM writes one-line results to the plutonium.csv or uranium.csv file, depending on the type of analysis. It writes one-line results to the FRAMcmd.log for the command line mode

analysis. FRAM v.6.1 adds extra information to the one-line results. Figure 14 shows a uranium CVS file displayed with Notepad.

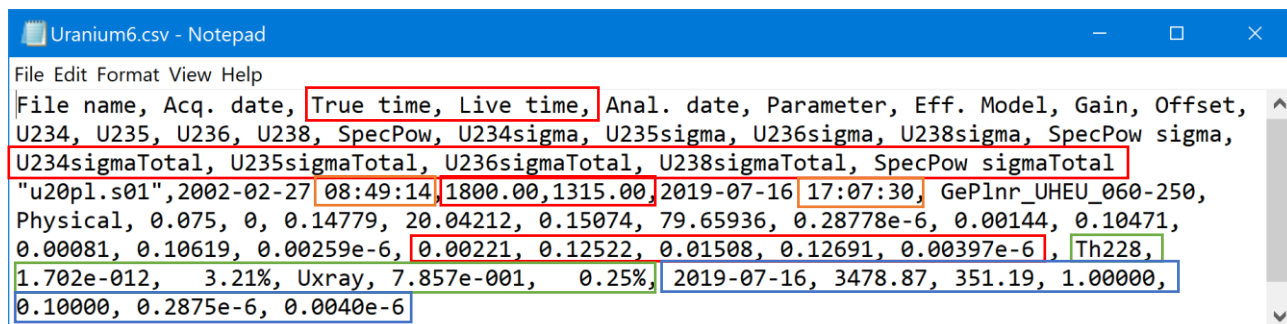


Figure 14. An example of a Notepad display of part of a uranium CVS file. The colored boxes show the added information from version 6.1 as compared with version 5.

The colored boxes show the new information being added to the one-line results. The red boxes show the labels and values of the true time, live time, and total uncertainties of the results. The orange boxes show the time together with the acquisition date and analysis date. The green boxes show the results of the ratio of an isotope to the main element (either U or Pu). In the figure, the values shown are for Th-228/U and Ux-ray/U ratios. In the analysis, when the option “Previous calorimetric measurement” or “Previous neutron measurement” is selected, FRAM will display the total uranium or plutonium mass results and other related information in the output displays and one-line results. The blue boxes show the mass and other information when the “Previous calorimetric measurement” option is checked. The descriptions for the seven values in these boxes are as follows: date of calorimetric measurement, total mass in g, uncertainty of total mass in g, sample power in mW, uncertainty of sample power in mW, specific power in W/gU, and error of specific power in W/gU. The uncertainties in the green and blue boxes are the total uncertainties (displayed in the medium/long output).

12. Constrained efficiency curve shape

The physical model of the relative efficiency curve employed by FRAM is a function of the self-attenuation, attenuation from external absorber, detector efficiency, and correction factor. The correction factor is to correct for the different detector efficiency and the inexactness of the internal and external attenuation due to the imprecise absorption coefficients of the nuclear material and the absorbers. The equation for the correction factor is the modified Hoerl function:

Correction factor $CF = E^b * c^{1/E}$ where E is energy, b and c are some variables.

In some measurements, such as those of the large UF₆ cylinders, where the container thicknesses are about the same and all the materials are infinitely thick, the efficiency curve for every measurement will be the same or very similar. In that case, it may be better if the efficiency curve can be fixed. FRAM v.6.1 adds two new parameters to help constrain the efficiency curve shape:

- i. *phy_eff_b* double fix the value b of E^b of the physical efficiency curve.
- ii. *phy_eff_c* double fix the value $\ln(c)$ of $c^{1/E}$ of the physical efficiency curve.

Together with the fixed or narrow ranges of the nuclear material and absorbers' thicknesses, the efficiency curve shape can be forced or constrained to some shape.

Due to the broad resolution and/or large tail of the LaBr₃ and CZT detectors, it is difficult to properly fit the regions and obtain correct areas of all the peaks. One or two bad peaks can negatively affect the efficiency curve and invalidate the curve. Therefore, it is useful and effective to constrain the efficiency curve shapes of the MRGS detectors to some general shape by setting the variables *b* and *c* to some values.

13. Pu242/U236 Declared date

When the “Measure | Analyze | Pu242/U236 by operator entry” option is selected, the “Declared date” text box will appear, allowing the declaration date to be entered. If the Declared Date text box is left blank, the date will be the date the spectrum is measured. Figure 15 shows an example of the Pu242/U236 operator entry option.

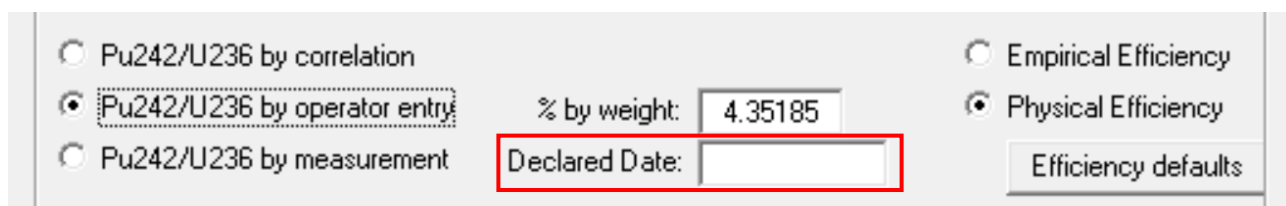


Figure 15. An example of Pu242/U236 by operator entry.

14. Date and time for fresh uranium analysis

For fresh uranium analysis, FRAM can correct for the non-secular equilibrium of uranium if the chemical separation date is known. For very fresh uranium, the correction is very large and can bias significantly if the separation time is off even a few hours. This leads to the need for the separation time, in addition to the date. Figure 16 shows an example of the Fresh Uranium analysis.

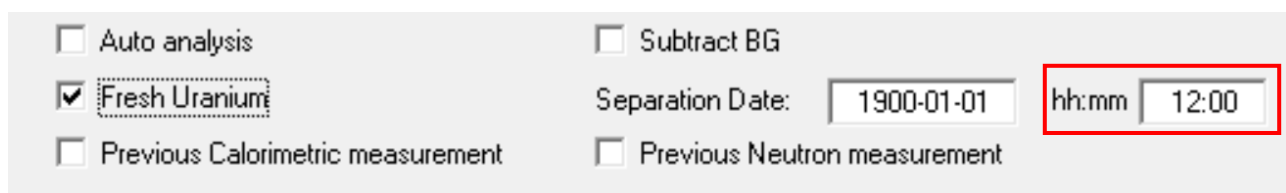


Figure 16. An example of Fresh Uranium analysis.

15. Change to keV dependent for FWHM and peak shape formulas

The FWHM and tail formula in the “Fitting Parameters” dialog box are changed from channel dependent to keV dependent to make them independent of energy calibration. Figure 17 shows the Fitting Parameters dialog box of version 5.

Edit Fitting Parameters -- Pu_Plnr_060-230

Default Energy Calibration

Gain (KeV/Ch) = Offset (keV) =

☐ Fixed

Default FWHM Constants

$fw\text{hm}(ch) = \sqrt{A1 + A2 \cdot E + A3/E}$

A1 (ch²) = A2 (ch²/KeV) = A3 (ch²*keV) =

☐ Fixed

Default Tailing Constants

$\text{tail}(ch) = H \cdot \exp[(T1 + T2 \cdot E) + (T3 + T4 \cdot E)(ch - x0)] \cdot [1 - \exp(-C \cdot (ch - x0)^2)]$

T1 = T2 = T3 = T4 =

☐ Fixed

Description:

Last modified on 06-Mar-2013

OK Cancel

Figure 17. Fitting Parameters dialog box of FRAM v.5.

A gamma-ray peak has its energy in keV and does not change, but its position in channel will change if the energy calibration changes. Therefore, it is clear that a change in the energy calibration would change the parameters A1–A3 of the FWHM formula and T1–T4 of the Tail formula.

Figure 18 shows the Fitting Parameters dialog box of version 6.1.

Edit Fitting Parameters -- GePlnr_Pu_060-230

Default Energy Calibration

Gain (keV/Ch) = 0.075 Offset (keV) = 0

☐ Fixed

Default FWHM Constants

$fwhm(keV) = \sqrt{A1 + A2 \cdot E + A3/E}$

A1 (keV²) = 0.06 A2 (keV) = 0.002 A3 (keV³) = 0

☐ Fixed

Default Tailing Constants

$tail(i) = H \cdot \exp[(T1 + T2 \cdot E) + (T3 + T4 \cdot E) \cdot (E_i - E)] \cdot [1 - \exp(-C \cdot (E_i - E)^2)]$

T1 = -4 T2 = 0.005 T3 = 4 T4 = 0

☐ Fixed

Description: Pu Planar, Equilibrium, U235/Pu < 0.2, 60-230keV

Last modified on 2019-06-17

OK Cancel

Figure 18. Fitting Parameters dialog box of FRAM v.6.1.

The magnitudes of the FWHM and tail at different energies depend on the detector system and are independent of the energy calibration. The keV-dependent FWHM and tail allow the user to modify the energy, FWHM, and tail parameters independently.

16. Expanded low-energy tail

The shape of a gamma-ray peak in the spectrum is described by a central Gaussian component with a single exponential tail on the low-energy side of the peak:

$$Y(E_i) = H \exp[-\alpha(E_i - E)^2] + Tail(E_i),$$

where

$Y(E_i)$ is the net count at energy E_i ,

H is the peak height at the peak energy E ,

$\alpha = 2.77259/FWHM^2$ the peak width parameter, and the tailing parameter $Tail(E_i)$ is given by

$$Tail(E_i) = H \exp[(T_1 + T_2 E) + (T_3 + T_4 E)(E_i - E)] * [1 - \exp(-0.4\alpha(E_i - E)^2)].$$

Here, we have allowed both the amplitude and slope of the tailing function to be a function of energy. In practice, we did not include T_4 in the fit in all previous versions of FRAM. That is equivalent to setting T_4 to zero and reducing the number of unknowns to three. For CZT spectrum, due to the large low-energy tail, the fit with three parameters T_1 , T_2 , and T_3 does not work well. We therefore programmed v.6.1 to include T_4 in the fit, and that apparently improves the fit of the CZT peaks.

When the command *expand_tail_model* in the Application Constants is set to *TRUE* then FRAM will include T_4 in the fit. For HPGe or LaBr₃ spectra, the low-energy tail is small, and the inclusion of T_4 in the fit will not improve the fit much but will make the fit less robust. It is suggested that for those spectra, one should not use the expanded tail model.

17. True physical efficiency model for heterogeneous material

In FRAM v.5, the physical model efficiency curve is represented by

$$RE = \left[\frac{(1 - \exp(-\mu_0 x_0))}{(\mu_0 x_0)} \right] * [\exp(-\mu_1 x_1) * \exp(-\mu_2 x_2) * \exp(-\mu_3 x_3)] * [A_i] * \left[\exp\left(\frac{c_j}{E}\right) \right] * [Detector Efficiency] * [Correction Factor].$$

The first term corresponds to self-transmission within the sample. The next three terms account for the transmission through as many as three different materials interspersed between the sample and the detector; A_i is the activity of isotope i ; $\exp(c_j/E)$ is the term for isotopic heterogeneity of isotope j ; *Detector Efficiency* is a generic detector efficiency parameterized in the code; and *Correction Factor* corrects for variations of the actual detector efficiency and the transmissions of the gamma rays through the nuclear material and the external absorbers from that specified in the model.

In FRAM v.6.1, we added the equation

$$RE = \left[\frac{(1 - \exp(-\mu_0 x_j))}{(\mu_0 x_j)} \right] * [\exp(-\mu_1 x_1) * \exp(-\mu_2 x_2) * \exp(-\mu_3 x_3)] * [A_i] * [Detector Efficiency] * [Correction Factor],$$

where x_j corresponds to the thickness of the nuclear materials representing different efficiency curves.

For nuclear material representing the first efficiency curve, x_j is x_0 , and the equation is the same as before. For nuclear material representing the second (or higher) efficiency curve (heterogeneity), x_j corresponds to the thickness of that nuclear material. The absorption coefficients for all the nuclear materials are assumed to be the same in this model.

The command for the true heterogeneous model is *true_hetero_model*. The default is *TRUE*. To revert to the $\exp(c_j/E)$ model, set the command to *FALSE*. The true heterogeneous model allows the

efficiency curve to cross the K-edge so we can have the heterogeneous parameter set employing the X-ray energy region of the analysis.

This command does not have an effect on the empirical heterogeneous efficiency model. The empirical heterogeneous efficiency model always uses the 1/energy factor.

18. List file in command line mode

A new command in the command line mode has been added. The command is `/L filelist`, where *filelist* is a text file containing rows of one-line commands such as `"/i inputfile.spe /p param"`. This command allows analysis of multiple files in command line mode. Figure 19 shows an example of an input list file named *FRAMcmdListFileTest.txt*.

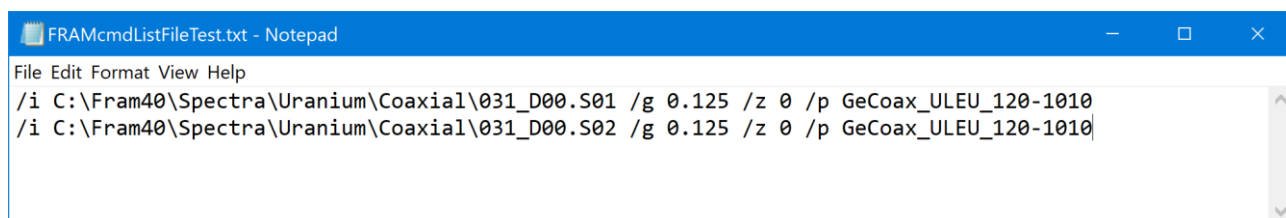


Figure 19. Example of an input list file named *FRAMcmdListFileTest.txt*.

Running command line mode *FRAM.exe /L FRAMcmdListFileTest.txt* will analyze the two spectra specified in the list file and write the results to the *FRAMcmd.log* file. Figure 20 shows the content of the *FRAMcmd.log* file after the analysis.

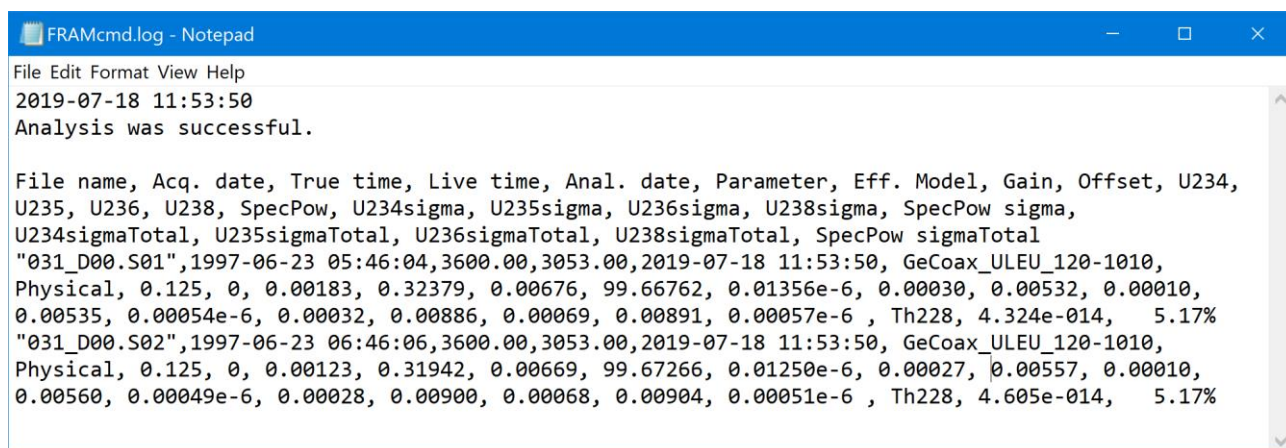


Figure 20. Content of the *FRAMcmd.log* file after running the *FRAMcmdListFileTest.txt* in Figure 19.

19. Help | Help topics

In FRAM v.5, clicking on the Help | Help topics button opens the Microsoft WinHelp FRAM.HLP file. This type of file was supported by Windows 3.0 through Windows XP. Its support was removed for Windows Vista and later. For Windows Vista and later Windows platforms, the Help inside the

FRAM program does not work; the users have to open a separate Microsoft Compiled HTML Help file.

In FRAM v.6.1, the user can now open the Microsoft Compiled HTML Help file from inside the FRAM program.

20. Minor improvements/changes

There are a number of minor improvements and changes that need to be mentioned, or users may not notice them. Some of these improvements are simple and do not take much effort to complete; some are complex and take a significant amount of time to finish even though they may appear simple on the surface.

a. Real number for true time and live time

FRAM v.5 and earlier versions use whole numbers for the measurement true time and live time. FRAM v.6.1 uses real numbers for true time and live time.

b. Spectrum display option

When a file is open, if “Region,” “Ecal,” “Fcal,” or “Scal” was previously selected, the same selection will also apply to the new file; if another radio button was selected, “SpecOnly” will apply.

c. Change maximum FWHM rule

In FRAM v.5.2, when a peak FWHM is greater than 32 channels (default), FRAM will ignore the peak, and it will not be fitted. FRAM v.6.1 introduces the parameter *max_fwhm_ch* to set the maximum channel such that FRAM will ignore the peak if the FWHM exceeds it.

d. Display results

The displays/prints of the isotopic results are changed from four decimal points to five decimal points.

e. Efficiency curve fit warning

The command *eff_reduced-chisq* will send a warning if the reduced chi-square of the efficiency curve fit is larger than the set value.

f. Background function fit

FRAM v.6.1 adds the quadratic step background. This background requires four background regions like that of the bi-linear step background.

g. Quadratic background

The quadratic background is forced to concave downward.

D. COMMAND-LINE MODE

FRAM version 6.1 adds several commands used in the command line mode operation.

- **/b BG file** Subtract the background peaks using a background spectrum. The default directory is **C:\FRAMdata\Spectra**.
- **/f Input file format** Specify the file format for the input file. In FRAM v.5, if **/f** is not present, the input file must have the extension chn, spc, mca, cnf, spe, or txt, and the file format will be automatically assigned to Ortec chn, Ortec spc, Canberra S100, Canberra Cam, IAEA MMCA, or FRAM Ascii, respectively. In version 6.1, if **/f** is not present, FRAM will read the file using the Auto format.
- **/j N42 spectrum #** The spectrum number of the N42 file. It is used together with **/f n42** command. The default value is 1.
- **/L List file** List file containing rows of one-line commands such as “/i inputfile.spe /p param”. This command allows analysis of multiple files in command line mode.

E. APPLICATION CONSTANT NAMES

FRAM version 6.1 adds a number of commands for the Application Constants of the FRAM parameter set as shown below.

Command	Type	Default	Description
Med_Resolution	BOOL	FALSE	Control the analysis algorithm. The Medium Resolution analysis is used for medium resolution spectra such as those of the LaBr ₃ detector or large CZT detector.
U234_corr	BOOL	FALSE	Turn on the ²³⁴ U correlation. The correlation has the form $\frac{^{234}\text{U}}{^{235}\text{U}} = a \frac{^{235}\text{U}}{^{238}\text{U}} + b, \frac{^{234}\text{U}}{^{235}\text{U}} \leq c$.
U235toU238slope	Real	0.000232	The correlation constant <i>a</i> in the expression for the ²³⁴ U correlation.
U235toU238zero	Real	0.007	The correlation constant <i>b</i> in the expression for the ²³⁴ U correlation.
U234toU235max	Real	0.012	The upper limit of the ²³⁴ U / ²³⁵ U ratio.
Pu238_corr	BOOL	FALSE	Turn on the ²³⁸ Pu correlation. The correlation has the form $\frac{^{238}\text{Pu}}{^{239}\text{Pu}} = c \left(\frac{^{240}\text{Pu}}{^{239}\text{Pu}} \right)^d$.
Pu240toPu239const	Real	0.274	The correlation constant <i>c</i> in the expression for the ²³⁸ Pu correlation.
Pu240toPu239exp	Real	3.	The correlation constant <i>d</i> in the expression for the ²³⁸ Pu correlation.
Pu240_corr	BOOL	FALSE	Set the boundaries such that if the measured ²⁴⁰ Pu activity is less than the lower bound, it will be set equal to the lower

			bound, and if it is greater than the upper bound, it will be set equal to the upper bound. The correlation has the form $\frac{{}^{240}\text{Pu}}{{}^{239}\text{Pu}} = a \left(\frac{{}^{241}\text{Pu} + {}^{241}\text{Am}}{{}^{239}\text{Pu}} \right)^b.$
AmPu241toPu239const	Real	1.0807	The correlation constant a in the expression for the ${}^{240}\text{Pu}$ correlation.
AmPu241toPu239exp	Real	0.5	The correlation constant b in the expression for the ${}^{240}\text{Pu}$ correlation.
Pu240_fractionlimit	Real	0.5	Set the bounds. The lower limit is set to be $(1 - \text{Pu240_fractionlimit})$ times the activity determined from the correlation, and the upper limit is $(1 + \text{Pu240_fractionlimit})$ times the correlated activity.
phy_eff_b	Real	NULL	Fix the value b of E^b of the physical efficiency curve to constrain the efficiency curve shape.
phy_eff_c	Real	NULL	Fix the value $\ln(c)$ of $c^{(1/E)}$ of the physical efficiency curve to constrain the efficiency curve shape.
expand_tail_model	BOOL	FALSE	Fit the tail using parameter T_4 in addition to the parameters T_1 - T_3 .
max_fwhm_ch	Real	32.	Set the maximum channel such that FRAM will ignore the peak if the FWHM exceeds it.
eff_reduced-chisq double	Real	100.	Send a warning if the reduced chi-square of the efficiency curve fit is larger than the set value.
correct_u_sumpeaks	BOOL	TRUE	In FRAM v.5, this command corrects for the intensities of the ${}^{238}\text{U}$ peaks that undergo the sum peak problem in the analysis employing the high-energy region above the X-ray. In FRAM v.6.1, this command also corrects for the intensities of the intrinsic Th-231 X-ray peaks that undergo the sum peak problem in the analysis employing the X-ray region.

F. CONCLUSION

We have introduced the new features and improvements of FRAM version 6.1. FRAM v.5.2 was written using Microsoft Visual C++ 6, which runs on Windows XP but not Windows 7 or later Windows. The FRAM v.5 code created by Microsoft Visual C++ 6 can run on all Windows platforms, from Windows 95 to Windows 10. Despite the greatness of Microsoft Visual C++ 6, for sustainability, we have to upgrade our Windows platform and programing code.

Windows XP and later Windows 7 are gradually being replaced by Windows 10. Since Microsoft Visual C++ 6 cannot run on Windows 10, we have to write the FRAM v.6.1 using the Microsoft Visual C++ 2012 installed on a Windows 10 computer. The executable FRAM v.6.1 created from this Microsoft Visual C++ 2012 can run on only Windows XP, Windows 7, and Windows 10. It cannot run on the earlier Windows platform. We believe the source code for FRAM v.6.1 can also be compiled with the latest Microsoft Visual C++ 2019 with minimal modification. Its capability to be

compiled and linked using the latest Microsoft Visual C++ has significantly improved the sustainability, compatibility, and maintainability of the FRAM code.

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